



## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of:

Group Art Unit: 1712

CHRISTIAN HERZIG ET AL.

Examiner: Marc S. Zimmer

Serial No.: 10/521,376

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For: BRANCHED SILOXANE POLYMERS COMPRISING ALKENYL GROUPS AND USED AS ANTIMISTING ADDITIVES FOR SILICONE COATING COMPOSITIONS

Attorney Docket No.: WAS 0675 PUSA

DECLARATION OF DR. CHRISTIAN HERZIG

Commissioner for Patents  
United States Patent and Trademark Office  
Washington, D.C. 20231

Sir:

I, Dr. Christian Herzig, do hereby declare and state as follows:

1. I am a co-inventor of the claimed subject matter of U.S. Application Serial No. 10/521,376, and am familiar with the contents of the application, the Office Action recently received, the *Clark* reference, U.S. Patent 6,586,535, and the *Inokuchi* reference U.S. 5,527,841.

2. *Clark* discloses antimisting additives which are very different from those claimed in the present invention. The *Clark* antimisting additives are prepared by hydrosilylating a large excess of an at least tris[alkenyl]-functional polysiloxane with an organosilicon compound containing at least two silicon-bonded hydrogens. However, the resulting additives are difficult to employ in traditional coating systems for at least two reasons. First, the additives tend to gel, thus not producing a liquid product. The gel compositions are often not soluble in the remaining coating system components. Second, most coating systems employ bis[alkenyl]-functional organosilicon compounds. The addition

of antimisting additives dissolved in a considerable excess of a tris- or higher[alkenyl]-functional compound will significantly alter the crosslink density of the coating formulation, changing its cured properties as well.

3. In the present application, our antimisting additives are branched alkenyl-functional additives prepared by reacting an excess of an  $\alpha,\omega$ -bis[alkenyl]-functional organosilicon compound with an SiH-functional organosilicon compound containing three or more silicon bonded hydrogen atoms (SiH). These additives, in contrast to those of *Clark*, are liquids which show no tendency to gel. Moreover, because they are prepared in excess bis[alkenyl]-functional organosilicon compound, the presence of even very significant amounts of additive produces little change in the cured coating.

4. To illustrate the differences between the subject invention and *Clark*, reference may be had to subject invention Example 7 and Comparative Experiment 2 (page 29 of the application, and to a new subject invention Example 6a and new Comparative Experiment 3 presented below.

In Example 7, a mist suppressing additive is prepared in accordance with the subject invention and found to be a clear colorless liquid with a viscosity of 1940 mm<sup>2</sup>/s at 25°C. In Comparative Example 2, the  $\alpha,\omega$ -bis[vinyl]-functional PDMS of Example 7 was replaced by a tris[vinyl]-terminated branched organopolysiloxane, as taught by *Clark*. The ratio of alkenyl groups to SiH groups was in excess, at 3.5:1. The product gelled to a toluene-insoluble hard rubber, useless as an antimisting additive.

The following two examples further demonstrate the differences between the subject invention and *Clark*, as follows:

Example 6a

The procedure of Example 6 is repeated with the modification that 250 g of a linear  $\alpha,\omega$ -di(5-hexenyl)polydimethylsiloxane with an iodine number of 7.1 are used instead of the linear  $\alpha,\omega$ -divinylpolydimethylsiloxane. The linear  $\alpha,\omega$ -di(5-hexenyl)polydimethylsiloxane is reacted in the same way as in Example 6 with a 4.37 g of the copolymer from Example 6 containing 0.32% by weight of Si-bonded hydrogen giving a C=C/SiH ratio of 5.0.

A clear, colorless liquid is obtained which has a viscosity of 1080 mm<sup>2</sup>/s at 25°C. The product has a hexenyl equivalent weight of 4560 g/mol C=C. It is suitable as an antimisting additive for silicone coating formulations.

Comparative Example 3

The procedure of Example 6a is repeated with the modification that 207 g of a linear trimethylsiloxy-terminated copolymer having 157 dimethylsiloxy units and 4.2 5-hexenylmethylsiloxy units, and with an iodine number of 8.6, are used instead of the linear  $\alpha,\omega$ -di(5-hexenyl)polydimethylsiloxane. As in Example 6a, 4.37 g of the copolymer containing 0.32% by weight of Si-bonded hydrogen are reacted, giving a C=C/SiH ratio of 5.0.

In accordance with a procedure analogous to that of Example 6a, an increase in viscosity occurs very rapidly, with subsequent gelling of the reaction mixture to form a rubberlike mass, which is insoluble in toluene and is not suitable as an antimisting additive for silicone coating formulations.

The comparative Example employs a linear organopolysiloxane with at least three (4.2) 5-hexenyl groups, as discussed by *Clark* at column 10, lines 40 - 42, while the subject invention example, to be as comparative as possible, employs an  $\alpha,\omega$ -bis[hexenyl]-functional PDMS. The hexenyl compounds were used in excess in both preparations, at a ratio of 5 hexenyl to 1 SiH group. *Clark* prefers an at least 4.6:1 ratio, so this comparative

example is squarely within *Clark*'s teachings. As can be seen, the additive of Example 6a is a clear, colorless liquid with a viscosity of only 1080 mm<sup>2</sup>/s at 25°C, while the produce of Comparative Example 3 gelled to form a rubber-like mass insoluble in toluene, unsuitable for use in coating compositions.

5. I am also familiar with *Inokuchi*, but am not aware of any relationship it has to silicone release coatings or antimisting additives. The *Inokuchi* coatings are for lubricious but abrasion resistant coatings, employing components not used in preparing crosslinkable silicone release coatings. First the a) components of *Inokuchi* are not crosslinkable. Second, the epoxy-functional component (b) is more likely to increase adhesion rather than provide a decrease. Finally, the solid rubber particles of *Inokuchi* are entirely unsatisfactory for use as antimisting additives. The latter must be liquid, and soluble in the remaining system components in order to exert an antimisting effect. One skilled in the art would not look to *Inokuchi* for any assistance in either preparing crosslinkable silicone release coatings or antimisting additives for use therein.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code.

  
Dr. Christian Herzog

Dated: Dec. 7th, 2006

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